

FINAL REPORT

Proposal No NAG 2-1103

Title: Modeling and Data Analysis of SONEX Observations

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First year: \$107,344 **Second year:** \$104,493 **Third year:** \$108,322 **Total:** \$320,159

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Standard 3-year Research Proposal XXX

New Investigator Program Proposal _____

Authorizing Official: Janis L. Goddard
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Georgia Tech Research Corporation
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Overview Of The Proposed Research

Primary focus of the study is on the sources, photochemistry and transport of reactive nitrogen species, O₃, as well as other O₃ precursors such as NMHC and CO. Major objectives are: (1) Identify the major sources of NO_x and quantify the contribution of each source to the NO_x distribution in the free troposphere, particularly in the upper troposphere. (2) Understand the odd hydrogen photochemistry and the recycling of NO_x in the free troposphere. (3) Evaluate the photochemical production and destruction of O₃.

A combination of modeling and data analysis/interpretation approach is used. We use a three-dimensional model and a box model to simulate, analyze, and interpret the observations. Both models are needed because they are complementary and each has its advantages and limitations. Proven analytical techniques, including correlation of NO_x with various tracers and with different air masses, are applied to both observations and model results to deduce useful information toward accomplishing the objectives. In addition, the SONEX observations are compared and analyzed in a systematic way against other airborne experiments conducted over remote atmosphere, especially those from NASA GTE experiments, AASE I and II, and SUCCESS.

Summary of Major Research Findings

(a) The relationship among NO_y, O₃, N₂O, ultra-fine condensation nuclei (CN), and other trace gases in the upper troposphere and lower stratosphere observed during SONEX are analyzed with the goal to identify and quantify the sources of NO_y in the upper troposphere. We use N₂O to separate upper tropospheric air (N₂O > 312 ppbv) from stratospheric influenced air (N₂O < 312 ppbv) and focus our analysis to the upper tropospheric air. The distributions of NO_y and O₃ show remarkable similarity when they are plotted as a function of N₂O, both in the stratospheric influenced air and in most of the upper tropospheric air. The only difference between NO_y and O₃ is found in upper tropospheric air where a large number of data points (about 20% of the entire observations) have high values of both NO_y (average mixing ratio about 1 ppbv) and NO_y/O₃ (greater than 10×10^{-3}). The major sources contributing to these high NO_y values are found to be emissions from lightning and surface sources convected to the upper troposphere.

(b) We examine concurrent measurements of CN (diameter > 8 nm), NO, and NO_y in the upper troposphere over the North Atlantic during the SONEX Experiment (Oct.-Nov., 1997). High CN and NO_y observations are attributed largely to the enhancement in convective outflow. Using the ratio of NO/NO_y as a chemical clock, we estimate that dilution of convective high-CN plumes is rapid (on a time scale of < 2 days) and accounts for a large fraction of elevated CN concentrations above the background. We estimate that less than 7% of observed high-CN (> 10000 cm⁻³) plumes may be attributed to aircraft emissions. The contribution by aircraft emissions to upper tropospheric CN

concentrations is estimated to be significantly higher than 7% because aircraft plumes dilute much faster than convective plumes and hence are sampled less frequently.

Publications:

Liu, S. C., H. Yu, B. Ridley, Y. Wang, D. D. Davis, Y. Kondo, M. Koike, B. E. Anderson, G. W. Sachse, S. A. Vay, G. L. Gregory, H. Fuelburg, A. Thompson, and H. Singh, Sources of Reactive Nitrogen in the Upper Troposphere During SONEX, *Geophys. Res. Letters*, 26, 2441-2444, 1999.

Wang, Y., S. C. Liu, B. E. Anderson, G. W. Sachse, S. A. Vay, Y. Kondo, A. Thompson, and H. Singh Evidence of convection of a dominant source of condensation nuclei in the northern mid-latitude upper troposphere, *Geophys. Res. Letters*, 27, 369-372, 2000.

Crawford, J., D. Davis, J. Olson, G. Chen, S. Liu, H. Fuelberg, J. Hannan, Y. Kondo, B. Anderson, G. Gregory, G. Sachse, R. Talbot, A. Viggano, B. Heikes, J. Snow, H. Singh, and D. Blake, Evolution and Chemical Consequences of Lightning Produced NO_x Observed in the North Atlantic Upper Troposphere. *J. Geophys. Res.*, In press, 2000.

Koike M., Y. Kondo, H. Ikeda, B. E. Anderson, G. W. Sachse, D. Blake, S. C. Liu, H. B. Singh, A. Thompson, K. Kita, Y. Zhao, T. Sugita, R. E. Shetter, and N. Toriyama, Impact of aircraft emissions on reactive nitrogen over the North Atlantic Flight Corridor region, *J. Geophys. Res.*, 105, 3665-3677, 2000

Kondo Y., M. Koike, H. Ikeda, B. E. Anderson, K. E. Brunke, Y. Zhao, K. Kita, T. Sugita, H. B. Singh, S. C. Liu, L. Jeagle, A. Thompson, G. L. Gregory, R. E. Shetter, G. W. Sachse, E. V. Browell, and M. J. Mahoney, Impact of aircraft emissions on NO_x in the lowermost stratosphere at northern midlatitudes, *Geophys. Res. Letters*, 26, 3065-3068, 1999.